

# Conversion between Three and Two Dimensional Optical Waves utilizing Surface Plasmon Excitations in Attenuated Total Reflection Kretschmann Configuration

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Conversion between three and two dimensional optical waves has been investigated utilizing surface plasmon excitations in the attenuated total reflection (ATR) Kretschmann configuration. Surface plasmons (SPs), two dimensional optical waves, were excited when dye molecules on metal thin film were directly irradiated by laser beams in the Kretschmann configuration. Emission light, three dimensional optical wave through the prism, was observed in the Kretschmann configuration using various organic thin films. The emission properties coincided with resonant conditions of SP excitations in the ATR Kretschmann configuration and multiple surface plasmons were simultaneously excited. The intensities and the spectra strongly depended upon nanostructured separation between organic films and metal thin films and/or dye molecules on metal thin films. It is thought that the phenomenon due to multiple SP excitations is very useful for conversion between three and two dimensional optical waves.

Key words: surface plasmon excitation, attenuated total reflection, Kretschmann configuration, molecular thin film, emission light.

## 1. INTRODUCTION

Surface plasmons (SPs), two dimensional optical waves that are a coupling mode of free electrons and light, can be resonantly excited on metal surfaces in the Kretschmann and the Otto configurations by electromagnetic waves due to the total reflection of a p-polarized laser beam [1-3]. SPs propagate along the surfaces and accompany strong electromagnetic waves, evanescent waves, that decay exponentially away from the surfaces [1-3]. SPs are also utilized in near field optics (NFO) with high frequency electromagnetic waves localized in structures smaller than the wavelength [3]. When SPs are resonantly excited at some incident angles of the total reflection in the Kretschmann and the Otto configurations, the reflected light is attenuated and the incident light, three dimensional optical waves, is almost converted to SPs, that is, two dimensional optical waves [1-3].

Recently, emission light at a resonant angle region of SP excitations was observed through the prism in the attenuated total reflection (ATR) Kretschmann configuration, when metal ultrathin films on the prism or molecular thin films on metal films were directly irradiated from air by a laser beam, that is, reverse irradiation [3-6]. The emission properties corresponded to the resonant conditions of SPs in the Kretschmann configuration, and it is considered that multiple SPs were induced by means of excitation of molecular thin films in the reverse irradiation [7-9]. Similar emission light due to multiple SP excitations was also observed when the SP was resonantly excited at the resonant incident angle in the ATR method of the Kretschmann configuration [10, 11].

In this study, emission light properties due to multiple SP excitations have been investigated for various nanostructured molecular thin films using the ATR

method and the reverse irradiation in the Kretschmann configuration. Conversion properties between three and two dimensional optical waves were also described.

## 2. EXPERIMENTAL DETAILS

Various molecular thin films, arachidic acid (C20) Langmuir-Blodgett (LB) films, merocyanine (MC) LB films, rhodamine-B (RB) LB films and spin-coated polyvinylcarbazole (PVK) films with cyanine dye (CY) were deposited on microscopic cover glasses covered with vacuum evaporated silver (Ag) thin films. MC, RB and CY are photosensitizing organic dyes showing photoluminescence (PL). MC (NK2684), RB (NKX736) and CY (NK1533) were purchased from Hayashibara biochemical Lab., Inc. C20 has no optical absorption and is one of dielectric materials [6]. Details of LB films have been reported elsewhere [9]. CY at 10 wt% was dispersed in spin-coated PVK films and the thickness of the film used in this study was about 19 nm. The thickness of the evaporated Ag films was about 50 nm and used as the SP active layers.

Figures 1 (a) and (b) show the measuring system for the ATR and the emission light and the reverse irradiation method in the Kretschmann configuration, respectively. A complete description has been reported elsewhere [9-11]. In the ATR measurement, reflectance intensity to incident one, that is, the ATR signal was measured as a function of the incident angle,  $\theta_i$ , of the laser beam. In this measurement, Ar<sup>+</sup> lasers ( $\lambda=488.0$  nm and 514.5 nm) and He-Ne lasers ( $\lambda=594.1$  nm and 632.8 nm) were used. In the reverse irradiation method, samples were irradiated at the vertical incident angle by p- and

s-polarized  $\text{Ar}^+$  laser beams at 488nm as shown in Fig.1 (b). Emission light through the prism was observed with and without a sharp cut filter below about 520 nm as a function of emission angle,  $\theta_e$ , where the light was observed [6-11]. Spectra of the emission light were measured at various emission angles [7-11]. Emission light through the prism was also observed at resonant SP excitations in the conventional ATR method as shown in Fig.1 (a).

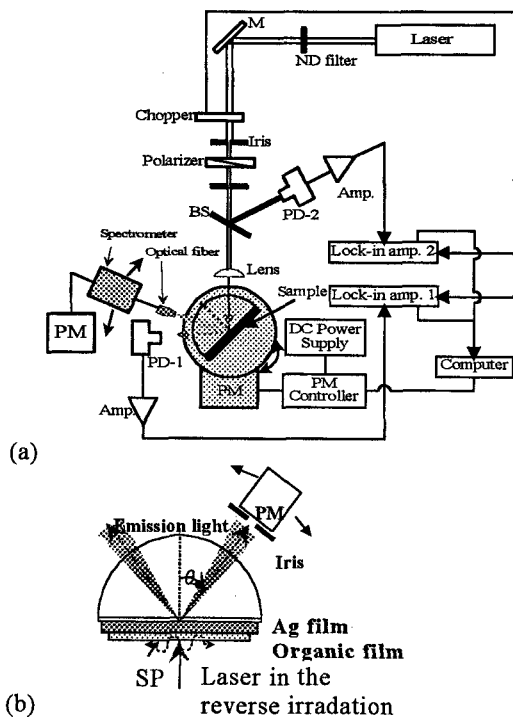


Fig.1 A measuring system for the ATR and the emission light (a) and the reverse irradiation method in the Kretschmann configuration (b).

### 3. RESULTS AND DISCUSSION

#### 3.1 ATR and emission properties

Figures 2 show ATR properties and the emission light for Ag film, Ag/C20 (6 layers)/MC (2 layers) and Ag/C20 (6 layers)/MC (8 layers) films using a p-polarized laser at 488 nm. The ATR properties in Fig.2 (a) have minima at the resonant angles ( $\theta_{SP}$ ) due to resonant excitations of SPs at 488 nm. Figures 2 (b) and (c) show emission light in the reverse irradiation of a p-polarized laser beam without and with a sharp cut filter, respectively. The emission light without the filter in Fig.2 (b) showed the peaks at about 44°, 50° and 60° that were the same angles as the resonant ones. The emission light contained mainly the wavelength of the laser. The emission light from the Ag films had only the laser wavelength of 488 nm and was not observed using the filter. However, the emission peaks with the filter in Fig.2 (c) were observed at about 44° and 47° for Ag/C20 (6 layers)/MC (2 layers) and Ag/C20 (6 layers)/MC (8 layers) films, respectively, and the spectra had the main peak at

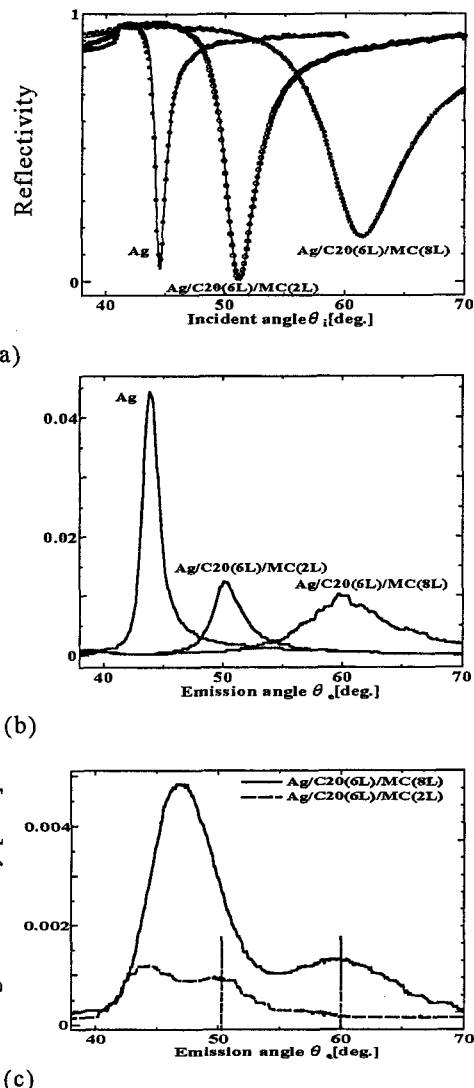


Fig.2 ATR properties (a) for Ag film, Ag/C20 (6 layers)/MC (2 layers) and Ag/C20 (6 layers)/MC (8 layers) films at 488 nm, and emission light in the reverse irradiation without (b) and with a sharp cut filter (c).

about 600 nm.

Emission light through the prism was also observed for Ag/RB (10 layers) film in the reverse irradiation of a p-polarized laser beam in the Kretschmann configuration. Figure 3 (a) shows spectra of the emission light at various emission angles. The spectra strongly depended on the emission angles. The peak wavelengths of the emission light became shorter as the emission angles increased. Each spectrum in Fig.3 (a) almost corresponded to a part of the broad PL spectrum of the RB LB films [7, 8]. Figure 3 (b) also shows emission spectra at various emission angles below the reflection angle of the  $\theta_{sp}$  in the conventional ATR measurement using the  $\text{Ar}^+$  laser at 488 nm when the SP was resonantly excited at 54.6° of the  $\theta_{sp}$  for the same Ag/RB (10 layers) sample, as shown in Fig.4. The property was very similar to one in Fig.3 (a) and the spectra also strongly depended on the emission angles.

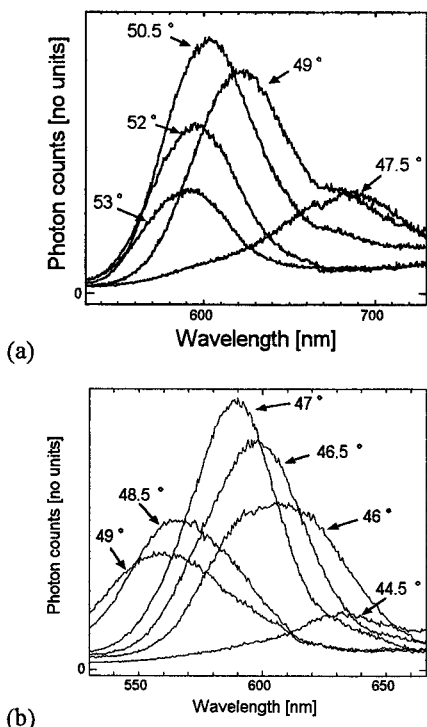


Fig.3 Emission spectra at various emission angles in the reverse irradiation (a) and due to the resonant excitation of the SP in the Kretschmann configuration (b) for the Ag/RB (10 layers) LB film.

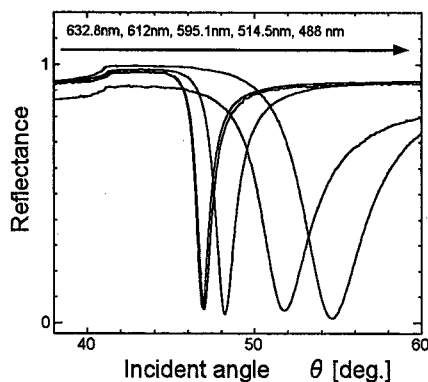


Fig.4 ATR properties at various wavelengths.

Figure 4 shows the ATR properties for the Ag/ RB film sample at various wavelengths using Ar<sup>+</sup> lasers ( $\lambda=488.0$  nm and 514.5 nm) and He-Ne lasers ( $\lambda=594.1$  nm, 612.0 nm, and 632.8 nm). The incident angles at the minimum reflectance that were the resonant ones of the SPs,  $\theta_{sp}$ , were 54.6°, 51.7°, 48.2°, 47° and 46.9° at 488.0nm, 514.5nm, 594.1 nm, 612.0 nm and 632.8 nm, respectively. The resonant angles  $\theta_{sp}$  shifted toward higher angle as the laser wavelengths became shorter. The relation between the  $\theta_{sp}$  and the wavelengths shows the dispersion property of SPs in the Kretschmann configuration of the prism/Ag/RB film sample. The dispersion property between the wavenumbers of the SPs,  $k_x$ , in the direction of the film surface parallel to the optical plane and the angular frequencies  $\omega$  of the laser wavelengths are calculated using the  $\theta_{sp}$  and the

wavelengths from the ATR properties at various wavelengths [7-11].

Figure 5 shows the dispersion property of the SPs calculated from the ATR properties in Fig.4. The property was indicated as open circles in Fig.5. The dispersion properties of the emission light in Figs.3 (a) and (b) were also calculated using the emission angles and the peak wavelengths, and are shown as full circles and triangles in Fig.5, respectively. These dispersion properties agreed well to one of the ATR measurements. These properties indicated that the emission light through the prism in the reverse irradiation and in the resonant excitation of the ATR measurement was caused by excitations of SPs in the ATR Kretschmann configuration.

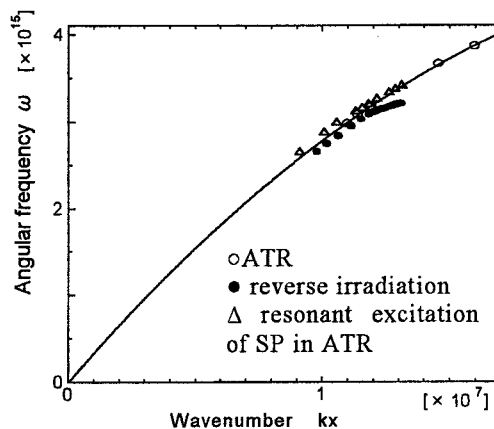


Fig.5 Dispersion properties of the SPs calculated from the ATR properties (○), from the emission light in the reverse irradiation (●) and from the emission light in the resonant excitation of SP in ATR (△).

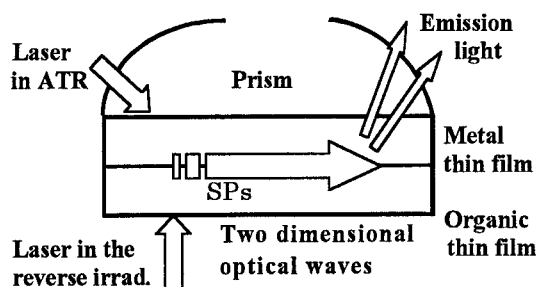


Fig.6 A schematic diagram of the emission phenomenon due to multiple SP excitations.

It is tentatively estimated that polarizations of organic dye molecules excited by the evanescent fields or the laser irradiation induce vibrations of free electrons at the metal surface producing multiple SPs, and the multiple SPs are converted to emission light at the resonant SP conditions in the Kretschmann configuration. Figure 6 shows a schematic diagram of the emission phenomenon due to multiple SP excitations. It is thought that the SP emission phenomenon involves the following processes: (1) multiple SP are induced by polarizations of excited organic dye molecules on metal thin films, (2) propagating on the metal surface, and (3) light that is

converted from propagating multiple SPs is emitted at the resonant SP conditions in the Kretschmann configuration due to film properties and/or roughness of the films. The phenomenon contains conversions from three to two dimensional optical waves and from two to three ones, and it is thought that emission light due to multiple SP excitations is very important for applications utilizing conversion between three and two dimensional optical waves.

### 3.2 Emission properties from CY dispersed PVK films in the reverse irradiation

Figure 7 shows emission light with the sharp cut filter as a function of emission angle for the CY dispersed PVK film irradiated by the laser beam of 488 nm at three polarized planes in the reverse irradiation. The emission peak by the p-polarized laser at 0° with the electric fields parallel to the observation plane was larger than that by 45° polarized one, and any considerable emission peak was not observed by the s-polarized one at 90°. Since CY dyes were dispersed uniformly in the spin-coated PVK and are rod-like, it is tentatively estimated that some dyes having the long axis parallel to the polarized plane of the laser were mainly excited by the reverse irradiation and the excited rod-like dyes induce anisotropic SPs propagating along the Ag surface in the observation plane.

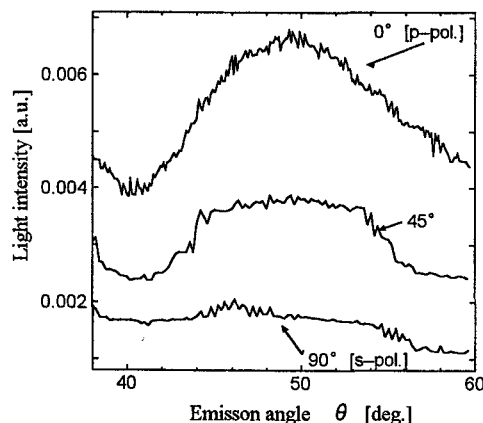


Fig.7 Emission light for the CY dispersed PVK film irradiated by the laser beam of 488 nm at three polarized planes in the reverse irradiation.

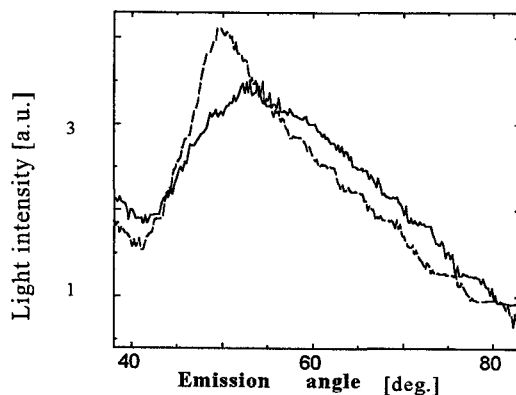


Fig.8 An example of the emission light before and after heat treatment at 100 °C for 20 min for the spin-coated PVK film with CY.

Since the emission phenomenon involves the three processes as shown in Fig.6, emission light can be modulated in the spectra and/or in the emission angle and will be utilized for a signal processing device if we can control whichever of the three processes. Figure 8 shows an example that the emission light by the p-polarized irradiation has been changed for the PVK film with CY before and after heat treatment at 100 °C for 20 min. The emission property in the emission angle varied after the heat treatment.

It is thought that the phenomenon due to multiple SP excitations is very useful for conversion between three and two dimensional optical waves.

## 4. COCLUSION

Emission light properties due to multiple SP excitations were investigated for various nanostructured molecular thin films using the ATR method and the reverse irradiation in the Kretschmann configuration. Conversion properties between three and two dimensional optical waves were also described. It is thought that the phenomenon is useful for applications.

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